

Figure 6-22. Short-term PM_{10} concentration time series for Missoula, MT, and Knoxville, TN.

season, Quarters 1 and 4. Regionally, the logarithmic standard deviation in the north-northwest is about 2.0 with pockets of high winter variability such as Salt Lake City, UT, and Missoula, MT. The lowest variability prevails over the warm season, Quarters 2 and 3, covering the southeastern and southwestern states. Over multistate regions in the southern states the summertime logarithmic standard deviation is below 1.5. This means that these areas are covered more or less uniformly by summertime PM_{10} , while the northern states are more episodic.

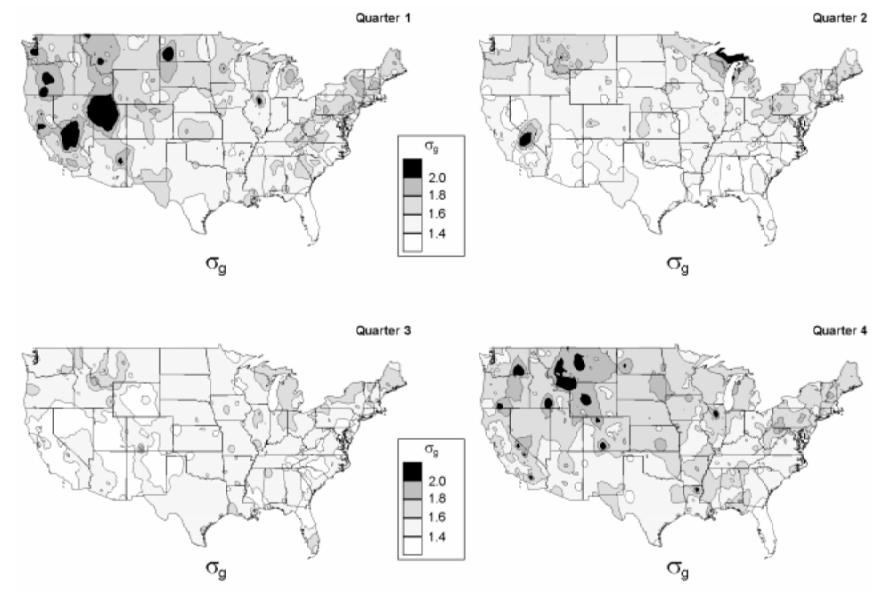


Figure 6-23. Geographic variation of the standard deviation, σ_g , of the lognormal distribution of PM $_{10}$ concentrations from the AIRS.

6.3.2.5 Aerometric Information Retrieval System PM_{2.5} Concentrations

The mass concentration of fine particles in urban areas is not well known. Sampling and analysis of $PM_{2.5}$ is limited by small number of stations (<50), sampling period restricted to few years, and different, non-standard sampling equipment was utilized for $PM_{2.5}$

The yearly average AIRS PM_{2.5} concentrations are shown in Figure 6-24. Figure 6-24 also shows the location and magnitude of PM_{2.5} concentrations from measurements of IMPROVE/NESCAUM monitoring networks. The fine particle data from the IMPROVE/NESCAUM show a pattern of high concentrations (> 15 μ g/m³) occurring over the eastern United States. This pattern of nonurban fine particle concentrations was discussed in Section 6.3.1.

6.3.2.6 Other National Surveys

A summary of urban PM_{10} , $PM_{2.5}$, PMCoarse at eight urban areas, Birmingham, AL, Buffalo, NY, Houston, TX, Philadelphia, PA, Phoenix, AZ, Pittsburgh, PA, Rubidoux, CA, and Steubenville, OH was reported by Rodes and Evans (1985). The overall ratio of the PM_{10} to Total Suspended Particulate (TSP) was 0.486. The relationships between PM_{10} and the 15 μ m fraction (IP) are linear for all sites. With exception of Phoenix, AZ, and Houston, TX, $PM_{2.5}$ exceeded the PMCoarse mass concentration in all six urban areas.

Spengler and Thurston (1983) reported PM concentrations in six U.S. cities: Portage, WI, Topeka, KS, Kingston, TN, Watertown, MA, St. Louis, MO, and Steubenville, OH, using dichotomous virtual impactors in the two size ranges, $PM_{2.5}$, having $d_p < 2.5 \mu m$, and coarse particle mass with $2.5 < d_p < 15 \mu m$. All six cities displayed a seasonal trend of higher summertime and lower wintertime concentrations. Figure 6-25 displays the monthly mean concentrations ($\mu g/m^3$) for PM_{15} (inhalable PM or IP), $PM_{2.5}$ (fine mass), PM_{15} - $PM_{2.5}$ (coarse mass), and sulfate (as ammonium sulfate) for the six cities. The seasonal pattern for $PM_{2.5}$ is pronounced for these cities, especially St. Louis, Portage, and Harriman, where the July monthly mean $PM_{2.5}$ concentration is 60%, 60%, and 40% higher than the January monthly concentration in each city, respectively. Sulfate has been noted to have similar variations in other locations (Altshuller, 1980).

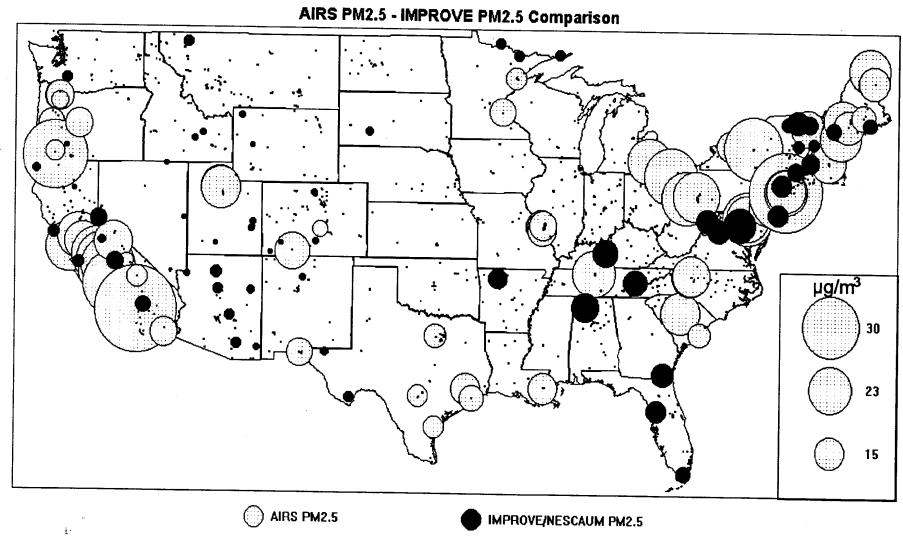


Figure 6-24. Annual PM_{2.5} concentration pattern obtained from IMPROVE/NESCAUM and AIRS networks.

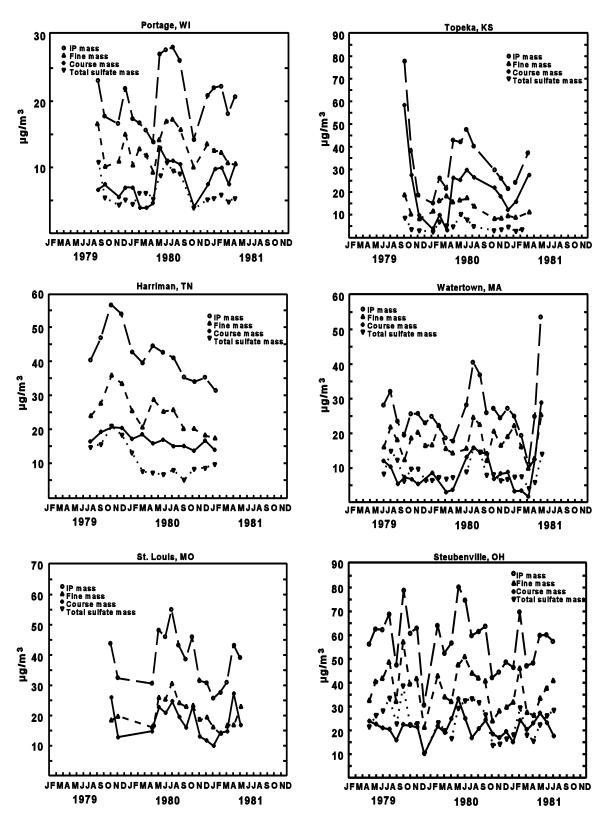


Figure 6-25. Monthly mean concentrations in micrograms per cubic meter of PM_{15} (IP, inhalable mass), $PM_{2.5}$ (fine mass), coarse mass (PM_{15} - $PM_{2.5}$), and total sulfate as (NH_4)₂SO₄ in Portage, WI; Topeka, KS; Harriman, TN; Watertown, MA; St. Louis, MO; and Steubenville, OH.

The quartz content and elemental composition of aerosols, collected in dichotomous samplers in selected sites in the EPA Inhalable Particulate network, were reported by Davis et al. (1984). For all network sites, an average of only 4.9 weight per cent of the coarse particle mass and 0.4 weight per cent of the fine mass consisted of quartz. Continental interior sites show the highest average quartz content as well as the greatest variability. The coastal regions and eastern interior sites reveal the lowest quartz concentrations. The complete X-ray spectra from some samples in Portland, OR, show that Si comes primarily from minerals such as feldspars, where the Si in the Buffalo, NY aerosols comes from quartz.

6.3.3 Comparison of Urban and Nonurban Concentrations

Seasonal maps of the AIRS PM₁₀-IMPROVE/NESCAUM PM₁₀ spatial concentrations are given in Figure 6-26. In evaluating the subsequent comparisons of the differences between AIRS and IMPROVE/NESCAUM spatial concentrations possible sampling biases and differences in sampling equipment and monitoring protocols may be significant. In addition, the differences in geographical location between the stations for the two networks also can influence the reliability of these comparisons. The AIRS PM_{2.5} concentrations everywhere exceed their adjacent IMPROVE/ NESCAUM concentrations. The highest AIRS PM_{2.5} are reported over the eastern urban industrial centers, such as Philadelphia and Pittsburgh, where the concentrations of 20 to 30 μ g/m³ exceed the nonurban PM_{2.5} by a factor of 2 to 3. However, the excess urban PM_{2.5} concentrations are evidently confined to the immediate vicinity of urban centers. This indicates that over the eastern United States a regionally homogeneous background of PM_{2.5} concentration exists that has smooth spatial gradients. Superimposed on the smooth regional pattern are local hot-spots with excess concentrations of factor of 2 to 3 that are confined within a few miles of urban industrial centers. The regional homogeneity is an indication that the eastern U.S. PM_{2.5} is composed of a secondary aerosol that is produced several days after the emission of its gaseous precursors. Similar results have been discussed for SO₄²⁻ since the 1970's (Altshuller, 1980). The excess PM_{2.5} concentration in urban centers suggests that primary emissions such as automobile exhaust and heating furnaces are responsible for much the urban PM_{2.5} hot-spots.

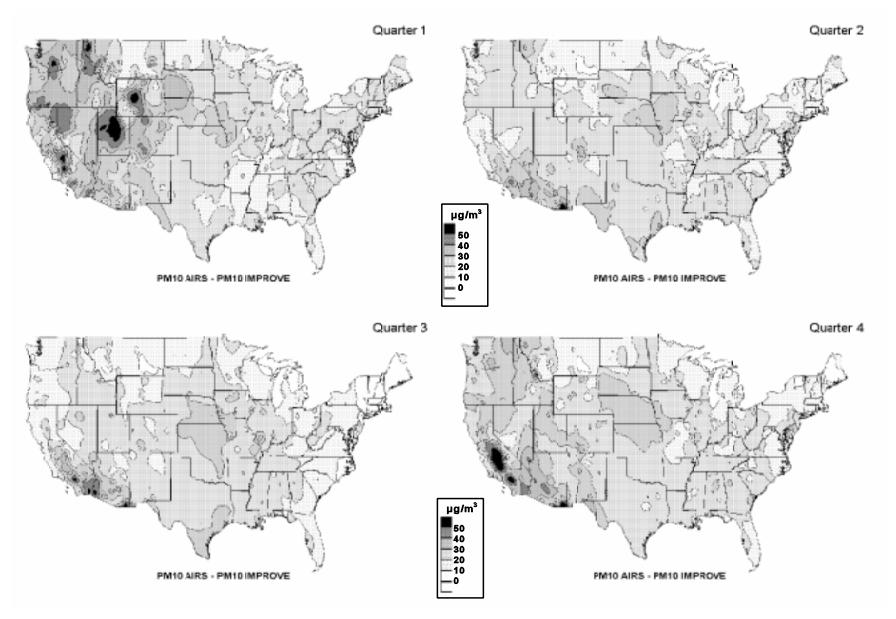


Figure 6-26. Spatial maps of PM_{10} concentration difference between AIRS and IMPROVE/NESCAUM networks.

The reported AIRS $PM_{2.5}$ concentrations over the Pacific states are generally higher and average at 20 to 50 μ g/m³. This is 5 to 10 times higher than their companion IMPROVE $PM_{2.5}$ concentrations. The dramatic difference is attributable to the pronounced concentration differences between urban-industrial-agricultural centers that exist in mountainous air basins and the concentrations monitored at remote national parks and wilderness areas that are generally at higher elevations. However, it is fair to presume that the AIRS and IMPROVE $PM_{2.5}$ data sets represent the extreme of aerosol concentration ranges that exist over the western U.S. The challenging task of filling in the details (i.e., spatially and temporally extrapolating the aerosol concentrations over the rugged western United States) is discussed in further detail in later regionally and locally focused sections below.

Comparisons have been made of the seasonality of the urban (AIRS) concentrations relative to the nonurban (IMPROVE/NESCAUM) data. In Figure 6-27 the difference in PM₁₀, PM_{2.5}, and PMCoarse between AIRS and IMPROVE/NESCAUM sites, using all available data, is used to indicate the urban excess particle concentration compared to the rural concentration. No attempt has been made to evaluate the possible uncertainties in these difference values.

Nationally, the urban excess fine particle concentration ranges between $18 \,\mu g/m^3$ in December through February and $10 \,\mu g/m^3$ in April through June (Figure 6-27a). The urban excess coarse mass concentration ranges between 10 to $7 \,\mu g/m^3$. The sum of the fine and coarse national urban excess mass concentration is about $25 \,\mu g/m^3$ in the winter season, and $18 \,\mu g/m^3$ during the spring season. Hence, the nationally aggregated urban and nonurban data confirm that urban areas may have excess concentrations on the order of $20 \,\mu g/m^3$, and well over half is due to fine particles, particularly in the winter season.

The urban excess (AIRS-IMPROVE/NESCAUM difference) over the eastern United States (Figure 6-27b) shows fine particles excess of 8 to 12 μ g/m³, with higher value occurring during both winter and summer. The urban excess coarse mass in the eastern United States is only 5 to 8 μ g/m³, peaking during spring and summer. The sum of fine and coarse urban excess is 15 to 18 μ g/m³ throughout the year.

Urban Excess

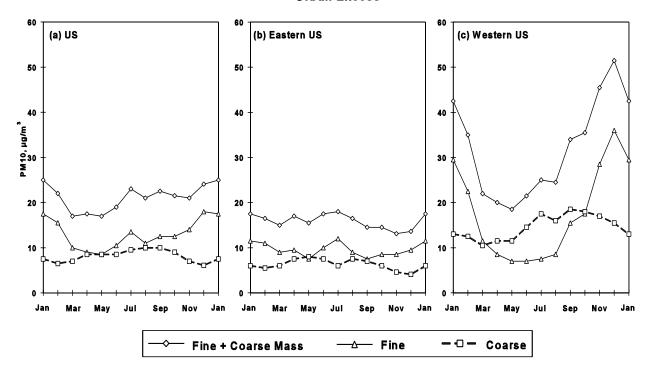


Figure 6-27. Urban excess concentrations (AIRS minus IMPROVE) for (a) the United States, (b) the eastern United States, and (c) the western United States.

The excess urban (AIRS-IMPROVE/NESCAUM) aggregated over the western United States is much more pronounced in magnitude and seasonality. The urban excess fine mass is about 30 μ g/m³ in November through January and drops to 8 to 10 μ g/m³ in April through August. The urban excess coarse mass is less in magnitude and seasonality 15 to 18 μ g/m³ in July through December, and 10 to 12 μ g/m³ in March through May. The sum of the urban excess fine and coarse mass is 40 to 50 μ g/m³ in November through January and about 20 μ g/m³ in the spring March through June. The urban AIRS and nonurban IMPROVE) networks in the western United States monitor aerosols differently because of different goals and mandates. The urban nonurban difference is such that the western nonurban concentrations contribute little to the much higher urban values, particularly in the winter season. On the other hand, the eastern urban sites are greatly influenced by the nonurban, regionally representative concentrations, particularly in the summer season.

6.4 REGIONAL PATTERNS AND TRENDS

This section describes the spatial, temporal, size, and chemical characteristics of seven aerosol regions of the conterminous U.S. The sizes and locations of these regions were chosen mainly on the basis of the characteristics of their aerosol pattern. The main criteria for delineating a region were (1) the region had to possess some uniqueness in aerosol trends, seasonality, size distribution, or chemical composition; (2) each territory of the conterminous United States had to belong to one of the regions; and (3) for reasons of computational convenience the shape of the regions were selected to be rectangular on unprojected latitude longitude maps. The resulting criteria yielded seven rectangular aerosol regions as shown in Figure 6-28. It is recognized that this selection is arbitrary and for future analysis additional regional definition criteria would be desirable. The limitations in the data bases of the two different networks discussed previously also apply to the subsequent discussion.

For sake of consistency and intercomparisons each region is described using maps delineating the spatial pattern and the sampling locations in the subsequent figures (Section a). For the figures showing AIRS monitoring results, Section b shows trends in average PM₁₀ concentrations and $\pm \sigma$. As discussed in Section 6.3.2.1 included in (b) are the results of two trend analyses. One of these uses the annual concentrations from all available stations in operation any time in the 1985 to 1994 period. The second approach uses the annual concentrations from only those stations operated continuously from 1985 to 1994, the long term coverage or trend stations. Section c show plots and correlations relating PM₁₀ and PM₂₅. Monthly AIRS concentrations (Section d) for a given region were computed by averaging all the available data for the specific month. In case of nonurban aerosol chemistry some regions only had two to four monitoring stations. The monthly nonurban PM_{2.5}, PMCoarse and PM₁₀ shown in the subsequent figures (Section b) over regions illustrate the relative seasonality of each aerosol type. The nonurban regional average chemical composition is presented as seasonal charts of chemical aerosol components as a fraction of the fine mass concentration (Section c). The role of some primary sources, such as coal and fuel oil combustion is indicated through seasonal charts of selenium (coal) and vanadium (fuel oil) trace metals (Section d). In addition, for each region figures will be provided showing shorter term variability of PM₁₀ concentrations and PM₁₀ urban excess concentrations.

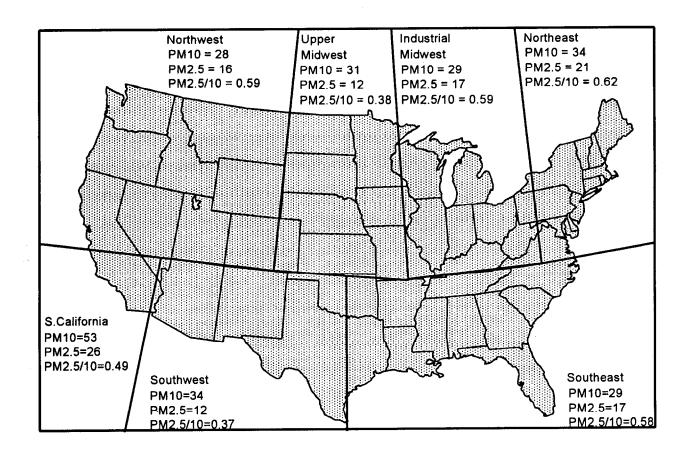


Figure 6-28. Aerosol regions of the conterminous United States.

6.4.1 Regional Aerosol Pattern in Eastern New York, New Jersey, and the Northeast

The Northeast aerosol region covers the New England states, including eastern Pennsylvania and eastern Virginia to the south (Figure 6-29a). In the Northeast, terrain features that significantly influence regional ventilation occur over the mountainous upstate New York, Vermont and New Hampshire. Throughout the year, the Northeast is influenced by Canadian as well as Gulf air masses. The region includes the Boston-New York megalopolis, as well as other urban-industrial centers. It is known that the Northeast is influenced by both local sources, as well as long range transport of fine particles and transformations of precursors to particles from other regions, as well as transport

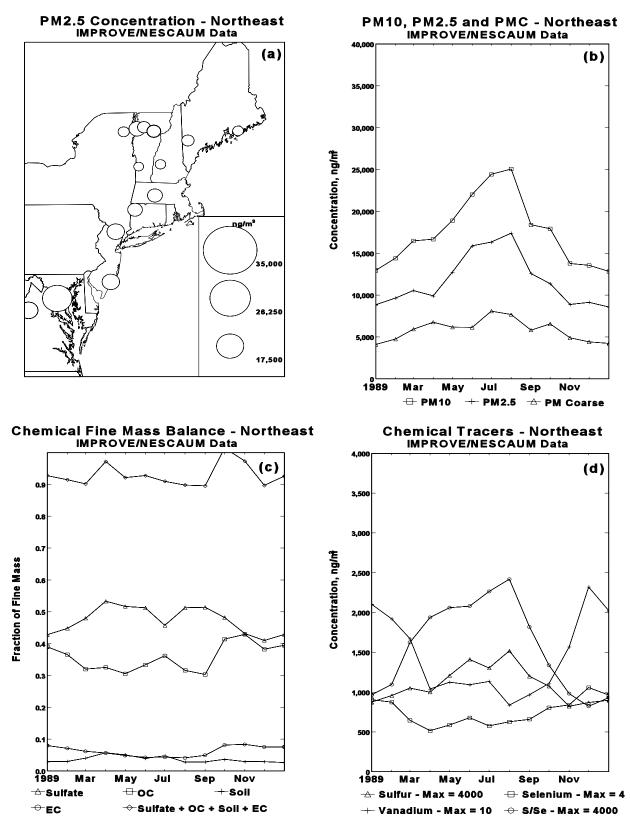


Figure 6-29. IMPROVE/NESCAUM concentration data for the Northeast: (a) monitoring locations; (b) PM_{10} , $PM_{2.5}$, and PMCoarse (PMC); (c) sulfate, soil, organic carbon (OC), and elemental carbon (EC) fractions; and (d) tracers.

and transformation of precursors in single and multiple urban plumes within the region (Chapter 3).

6.4.1.1 Nonurban Size and Chemical Composition in the Northeast

The summary of the nonurban aerosol chemical composition in the Northeast is presented in Figure 6-29c. The region has 14 monitoring sites, 8 of which are part of NESCAUM in upper New England. The geographic locations with respect to nearby urban areas vary from those sites within the northeast corridor to rural sites near the Canadian border.

The PM_{10} concentration exhibits a factor of two seasonal amplitude between $12 \ \mu g/m^3$ in the winter, and $25 \ \mu g/m^3$ in June and July (Figure 6-29b). About 60% of PM_{10} is contributed by fine particles throughout the year. The $PM_{2.5}$ also contributes to the summer-peaked seasonality.

Data from a two year fine particle network in the Northeast (Bennett et al., 1994) yielded a geometric mean concentration of $PM_{2.5}$ of 12.9 $\mu g/m^3$ and particulate sulfur (1.4 $\mu g/m^3$, equivalent to 4.2 $\mu g/m^3$ of sulfate), which is somewhat lower than other comparable rural data.

Sulfates are the most important contributors of the fine particle mass in the Northeast, particularly in the summer season when they account for half of the fine mass (Figure 6-29c). The regionality of sulfate in the northeastern U.S. has been dicussed for many years (Altshuller, 1980). The organic carbon ranges from 30 to 40%, with the higher percentages occurring in the fall and winter, September through January. In fact, during the late fall the contributions of sulfate and organic carbon are comparable at 40%. Fine particle soil is unimportant throughout the year (<5%). Elemental carbon, on the other hand, is somewhat more significant, particularly during the fall when it contributes about 10% of the fine mass. The sum of the above four nonurban fine particle aerosol components, account for over 90% of the measured fine particle mass throughout the year. These results would appear to indicate ammonium ion, hydrogen ion, nitrates, trace metals and sea salt are of minor importance in the northeastern U.S. fine particle chemical mass balance.